Evidence for the Sr_2RuO_4 intercalations in the $Sr_3Ru_2O_7$ region of the $Sr_3Ru_2O_7$ - Sr_2RuO_4 eutectic system

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Abstract. Although $Sr_3Ru_2O_7$ has not been reported to exhibit superconductivity so far, ac susceptibility measurements revealed multiple superconducting transitions occurring in the $Sr_3Ru_2O_7$ region cut from $Sr_3Ru_2O_7$ - Sr_2RuO_4 eutectic crystals. Based on various experimental results, some of us proposed the scenario in which Sr_2RuO_4 thin slabs with a few layers of the RuO_2 plane are embedded in the $Sr_3Ru_2O_7$ region as stacking faults and multiple superconducting transitions arise from the distribution of the slab thickness. To examine this scenario, we measured the resistivity along the *ab* plane (ρ_{ab}) using a $Sr_3Ru_2O_7$-region sample cut from the eutectic crystal, as well as along the *c* axis (ρ_c) using the same crystal. As a result, we detected resistance drops associated with superconductivity only in ρ_{ab}, but not in ρ_c. These results support the Sr_2RuO_4 thin-slab scenario. In addition, we measured the resistivity of a single crystal of pure $Sr_3Ru_2O_7$ with very high quality and found that pure $Sr_3Ru_2O_7$ does not exhibit superconductivity down to 15 mK.

1. Introduction

The Ruddlesden-Popper series of layered perovskites $Sr_{n+1}Ru_nO_{3n+1}$ is a fascinating research subject because the n=1 member Sr_2RuO_4 is now believed to be a spin-triplet superconductor [1, 2]. The n=2 member of this series $Sr_3Ru_2O_7$ is known to exhibit an enhanced Pauli paramagnetism with a metamagnetic transition [3, 4, 5]. Although crystals of $Sr_3Ru_2O_7$ are highly refined, the superconductivity in $Sr_3Ru_2O_7$ has not been discovered so far. Recently, $Sr_3Ru_2O_7$-Sr_2RuO_4 eutectic crystals were successfully grown [6] and surprisingly, multiple superconducting transitions were observed in the ac susceptibility measurements using a $Sr_3Ru_2O_7$-region sample cut from eutectic crystals [7], as presented in Fig. 1(a). However, it has been revealed that this superconductivity is not a bulk property of $Sr_3Ru_2O_7$ because these superconducting transitions are easily suppressed by small ac magnetic fields and no anomaly was observed in the specific heat [7]. The most plausible scenario of this superconductivity is that Sr_2RuO_4 thin slabs with a few layers of RuO_2 planes are embedded in the $Sr_3Ru_2O_7$ region and the multiple superconducting transitions arise from the distribution of the slab thickness.

In order to obtain additional evidence for the origin of the superconductivity observed in the $Sr_3Ru_2O_7$ region cut from eutectic crystals, we measured the resistivity along the *ab* plane (ρ_{ab}) of a $Sr_3Ru_2O_7$-region sample cut from the eutectic crystal, which we designate below as a eutectic $Sr_3Ru_2O_7$ sample, as well as along the *c* axis (ρ_c) of the same sample. In addition, we measured ρ_c of a pure (i.e. non-eutectic) $Sr_3Ru_2O_7$ sample with very high quality in order to clarify whether or not pure $Sr_3Ru_2O_7$ exhibits superconductivity at low temperatures.

2. Experimental

Resistivity was measured using a conventional four-probe method with an ac current. Single crystals of the $Sr_3Ru_2O_7$-Sr_2RuO_4 eutectic system and those of single-phase $Sr_3Ru_2O_7$ were grown by a floating-zone method. The size of the eutectic $Sr_3Ru_2O_7$ sample was approximately 1.5×0.7 mm^2 in the *ab* plane and 0.3 mm along the *c* axis. The results of ac susceptibility and specific heat measurements using this eutectic $Sr_3Ru_2O_7$ sample were reported in Ref. [7]. The dimensions of the pure $Sr_3Ru_2O_7$ sample are 0.44×0.28 mm^2 in the *ab* plane and 1.14 mm along the *c* axis. The ρ_{ab} measurements on the eutectic $Sr_3Ru_2O_7$ sample were performed down to 0.3 K with a ^{3}He cryostat (Oxford Instruments, model Heliox VL). After the ρ_{ab} measurements, we removed the electrical leads and attached another set of wires again on the same sample and performed ρ_c measurements down to 0.1 K with an adiabatic demagnetization refrigerator (Cambridge Magnetic Refrigeration, mFridge50). The ρ_c measurements using the pure $Sr_3Ru_2O_7$ sample were performed with a ^{3}He-^{4}He dilution refrigerator (Cryoconcept, model DR-JT-S-100-10) down to 15 mK. In this study, we used a cylinder of permalloy (Hamamatsu Photonics K.K., E989-28) in order to reduce remanent fields such as the earth field.

3. Results and Discussion

3.1. Resistivity of the eutectic $Sr_3Ru_2O_7$

Figure 1(b) shows temperature dependence of ρ_{ab} and ρ_c of the eutectic $Sr_3Ru_2O_7$ sample. The values of ρ_{ab} and ρ_c are approximately 1 μΩcm and 300 μΩcm at 1.5 K, respectively. The in-plane resistivity is nearly the same as those of pure $Sr_3Ru_2O_7$ crystals with very high quality [5]. This low value of the in-plane resistivity indicates that $Sr_3Ru_2O_7$ in the eutectic system crystallized with high quality. Also, macroscopic Sr_2RuO_4 domains in the eutectic system are high quality because its T_c is nearly 1.5 K [7], which is one of the best T_c of Sr_2RuO_4 reported. It is interesting that both Sr_2RuO_4 and $Sr_3Ru_2O_7$ spontaneously crystallize with high quality in this eutectic system.

In ρ_{ab} measurements, two clear resistance drops were observed at 1.05 and 1.32 K. These transition temperatures well coincide with those observed in the ac susceptibility [Fig. 1(a)]. However, in ρ_c measurements, no obvious transition was observed. These results are consistent with the Sr_2RuO_4 thin-slab scenario because they imply that superconducting inclusions embedded in the eutectic $Sr_3Ru_2O_7$ are too thin along the *c* axis to shortcircuit the current path along the interlayer direction. This behavior is in sharp contrast with ρ_c for the Sr_2RuO_4-Ru eutectic system, in which the emergence of the "3-K" superconductivity in the interface of Ru lamellae results in a large drop in ρ_c [8]. Although we observed two clear transitions in the ρ_{ab} measurement using the eutectic $Sr_3Ru_2O_7$ sample, ρ_{ab} does not become zero down to low temperatures. This non-zero resistivity implies that Sr_2RuO_4 inclusions in this eutectic $Sr_3Ru_2O_7$ sample do not completely form a path between the voltage contacts. The Sr_2RuO_4 inclusions are probably well separated in this sample. In some cases, eutectic $Sr_3Ru_2O_7$ samples exhibit zero resistivity (e. g., Ref. [9]), probably because Sr_2RuO_4 inclusions in such samples link a path between the voltage contacts. Now, on the basis of various experiments, we believe that the origin of the superconductivity observed in the eutectic $Sr_3Ru_2O_7$ sample is the presence of several monolayers of RuO_2 planes intercalated in $Sr_3Ru_2O_7$ as stacking faults. For example, two monolayers of RuO_2 planes intercalated in $Sr_3Ru_2O_7$ is schematically drawn in Fig. 1(c). In fact, such stacked monolayers of RuO_2 planes have been observed with a transmission electron microscope [9].

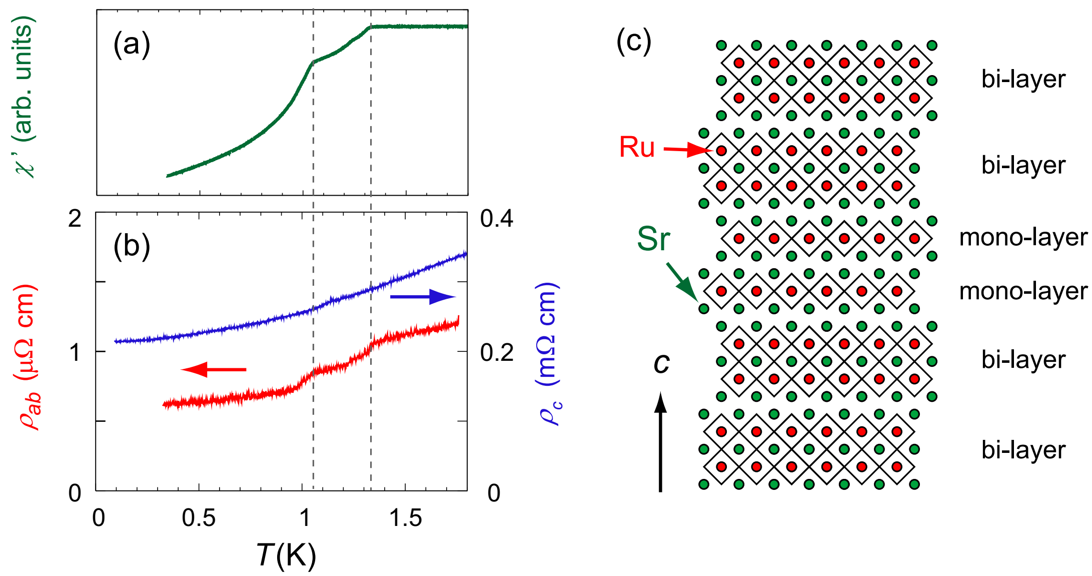

Figure 1. (a) Temperature dependence of the real part of the ac susceptibility for a eutectic $Sr_3Ru_2O_7$ sample with $\mu_0 H_{ac}$=0.58 μT and f=3011 Hz(sample 1 in Ref. [7]). (b) Temperature dependences of the resistivity along the *c* axis and that along the *ab* plane for the eutectic $Sr_3Ru_2O_7$ sample measured at f=89.1 Hz (I=0.5 mA-rms for ρ_{ab} and I=0.1 mA-rms for ρ_c). (c) A schematic image of two monolayers of RuO_2 planes (Sr_2RuO_4) intercalated in bilayers ($Sr_3Ru_2O_7$). Oxygens are located at the corner of the octahedra.

3.2. Resistivity of pure $Sr_3Ru_2O_7$ with high quality

We are also interested in the possibility of superconductivity in pure $Sr_3Ru_2O_7$. In order to examine the superconductivity in pure $Sr_3Ru_2O_7$, we consider it important (i) to use single crystals of single-phase $Sr_3Ru_2O_7$ with very high quality, (ii) to cool down the sample to sufficiently low temperature, and (iii) to perform measurements in zero field by excluding the geomagnetic field. Therefore, we measured ρ_c down to 15 mK using a single crystal of pure $Sr_3Ru_2O_7$ with the in-plane residual resistivity of 0.4 μΩcm, which is one of the highest-quality $Sr_3Ru_2O_7$ grown so far. In addition, by placing the sample in a cylinder of permalloy, we reduced the residual field to be lower than 0.1 μT.

Figure 2 shows temperature dependence of ρ_c of the pure $Sr_3Ru_2O_7$ sample. The ρ_c monotonically decreases with decreasing temperature and no anomaly indicating a superconducting transition was observed down to 15 mK. From this measurement, we conclude that pure $Sr_3Ru_2O_7$ does not become superconducting down to 15 mK.

4. Conclusion

We measured the resistivity along the *c* axis as well as along the *ab* plane of a eutectic $Sr_3Ru_2O_7$ sample. The resistance drops due to the multiple superconducting transitions were observed only for ρ_{ab}, but not for ρ_c. This result indicates that superconductors with thin thickness along the *c* axis are embedded in the eutectic $Sr_3Ru_2O_7$. Now, we are convinced that the origin of the superconductivity observed in the eutectic $Sr_3Ru_2O_7$ sample is the presence of Sr_2RuO_4 inclusions embedded in $Sr_3Ru_2O_7$ as stacking faults. In order to search for superconductivity in pure $Sr_3Ru_2O_7$, we also measured the resistivity along the *c* axis using a single crystal of best-quality pure $Sr_3Ru_2O_7$. However, no resistance anomaly associated with the superconducting transition was observed down to 15 mK. Therefore, we conclude that pure $Sr_3Ru_2O_7$ does not become superconducting down to 15 mK.

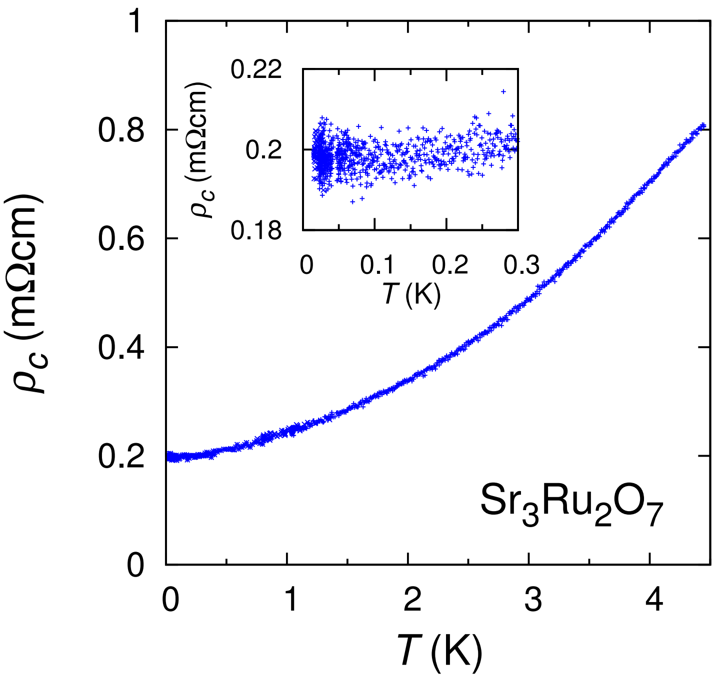

Figure 2. Temperature dependence of the resistivity along the *c* axis of the pure $Sr_3Ru_2O_7$ sample with very high quality measured at I=0.01 mA-rms with f=7 Hz without the geomagnetic field. The inset shows the low-temperature region below 0.3 K.

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